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Concentrations of PCDD/PCDFs and PCBs in spent foundry sands

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ABSTRACT

Approximately 10 million tons of spent foundry sand (SFS) are generated in the United States each year, and their beneficial use in agricultural and horticultural applications is being considered. Other studies have demonstrated that trace elements are low enough in sands from iron, steel, and aluminum foundries to allow their beneficial use. Because data were not available on polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, and polychlorinated biphenyls in SFSs, we analyzed representative spent sands from 10 foundries to assess the potential for these compounds to limit their use in soil-related applications. The total TEQ (toxicity equivalent) concentrations ranged from 0.01 to 3.13 ng TEQ kg⁻¹, with an average concentration of 0.58 ng TEQ kg⁻¹. These concentrations are within the range of natural background in soils.

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1. Introduction

Polychlorinated dibenzo-*p*-dioxin (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) are ubiquitous environmental contaminants (Wagrowski and Hites, 2000; Gambaro et al., 2005). They are non-polar, lipophilic, persistent in the environment, and bioaccumulate in the food chain. Unlike PCBs, PCDD/PCDFs were never intentionally manufactured, but are largely released into the environment during combustion processes (Fiedler, 1996) and have also been shown to form naturally in soils (Hoekstra et al., 1999). PCDD/PCDFs have been detected in emissions from municipal and hospital waste incinerators, steel mills, copper smelters, and automobiles (Hutzinger et al., 1985; Mininni et al., 2007; Choi et al., 2008). PCBs have been manufactured for use as dielectric fluids, lubricating fluids, hydraulic fluids, and flame retardants. Due to the toxicity and endocrine-disrupting properties of PCDD/PCDFs and PCBs (Birnbaum, 1995; Aylward et al., 1996; Aoki, 2001), much attention has been focused on monitoring these compounds in the environment and studying their effects on living organisms.

Sands are used by the foundry industry to create metalcasting molds, where the individual sand grains are typically bound using bentonite clays or resins. During the casting process, the molds can be exposed to temperatures as high as 1500 °C depending upon the metal being poured. The molding sands are reclaimed within the

foundry, but are eventually discarded in landfills when the sand grains are worn down by mechanical abrasion. In the United States, foundries generate about 10 million tons of spent foundry sand (SFS) annually. Efforts to divert the SFSs from landfills and encourage their beneficial use in manufactured soils and geotechnical applications are being supported by the US EPA (2007). Due to concerns about trace element and xenobiotic contamination, the USDA-ARS has undertaken efforts to characterize spent foundry sands and assess potential use risks in soil-related applications (Dungan, 2006; Dungan and Dees, 2007, 2009).

The objective of this study was to report concentrations of PCDD/PCDFs and PCBs in SFSs, as information of this type is lacking for many industrial byproducts in the literature. This information will be invaluable to state regulatory agencies that are considering measures that allow for the beneficial use of SFSs in agricultural and horticultural applications.

2. Materials and methods

2.1. Spent foundry sands

Spent sand samples were placed into 500-mL I-CHEM glass jars with Teflon-lined polypropylene closures (Chase Scientific Glass Inc., Rockwood, TN, USA), shipped to our laboratory in Styrofoam coolers with ice packs, and stored at 4 °C until processed. A total of 10 spent sands from iron, aluminum, and steel foundries were analyzed for PCDD/PCDFs and PCBs (Table 1). The spent sands consisted of green sands (bonded with sodium and/or calcium

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Table 1

Description of the spent foundry sands.

Sand	Metal poured	Molding sand	Core binder system and process
1	Aluminum	Green sand	Shell
2	Iron	Green sand	PU coldbox, PU hotbox
3	Iron	Shell	Shell
4	Aluminum	Green sand	PU no-bake, shell, core oil
5	Iron	Green sand	PU coldbox, PU hotbox
6	Aluminum	Green sand	Shell
7	Iron	Green sand	None
8	Steel	PU no-bake	PU no-bake
9	Steel	Green sand	PU coldbox, shell, resin/CO ₂
10	Steel	Green sand	PU no-bake, shell, core oil, resin/CO ₂

PU, phenolic urethane.

bentonite clay) and chemically bonded sands (i.e. shell and phenolic urethane no-bake). Shell is a molding process where a phenolic novolac film on sand is cured using a heated pattern, whereas a phenolic urethane no-bake is cured at room temperature using an acid catalyst (Gardziella et al., 2000).

2.2. PCDD/PCDF and PCB analysis

The SFSs were processed and analyzed for PCDD/PCDFs by US EPA Method 1613 (EPA, 1994) and modified to include dioxin-like PCBs (IUPAC Nos. 77, 126, and 169). PCB-81 and mono-ortho-substituted PCBs were not analyzed in this study. This procedure has previously been reported for feed and wood samples (Huwe and Smith, 2005) and is briefly summarized here. The spent sands (10 g) were air dried overnight, extracted three times by sonication in toluene:acetone (70/30, v/v) for 1 h, dried with sodium sulfate, and purified on a PowerPrep automated dioxin cleanup instrument

(Fluid Management Systems, Waltham, MA, USA) using multiphase silica gel, basic alumina, and carbon cartridges. The chemical standards used for the analysis were purchased from Wellington Laboratories (Guelph, ON, Canada). Quality assurance and control were maintained through the use of blanks, spikes, and surrogates. Recovery of PCDD, PCDF, and PCB spikes were within 15% of the true value, except for PCB-77 and OCDD which were within 25%. The ¹³C-labeled surrogate recoveries ranged from 40% to 149%. All recoveries are within the quality criteria established by USEPA Method 1613.

Toxic equivalency values (TEQs) were calculated by summing the products of each congener concentration and its World Health Organization (WHO) 2005 toxic equivalency factor (TEF) (Van den Berg et al., 2006).

3. Results and discussion

The concentrations of the PCDD/PCDFs and PCBs in the SFSs, expressed as ng kg⁻¹, are presented in Table 2. Except for 1,2,3,7,8,9-HxCDF, the tetra (T), penta (Pe), hexa (Hx), hepta (Hp) and octa (O) congeners of PCDD and PCDF were detected above detection limits, but not in all SFSs. Concentrations of the PCDD congeners ranged from <0.01 to 44.8 ng kg⁻¹, with 1,2,3,4,6,7,8,9-OCDD being found at the highest concentration in all of the spent sands. 2,3,7,8-TCDD (and 1,2,3,7,8-PeCDD) have been assigned a TEF of 1, whereas the TEF for OCDD is 0.0003 (Van den Berg et al., 2006). 2,3,7,8-TCDD, with concentrations ranging from <0.01 to 0.14 ng kg⁻¹, was detected in only 50% of the foundry sands. In a study conducted by Schmid et al. (2005), background concentrations of 2,3,7,8-TCDD in various agricultural, grassland, and forest soils ranged from <0.05 to 0.7 ng kg⁻¹ (Table 3). Similar results were also obtained in a Norwegian soil survey for TCDD and PeCDD, HxCDD, HpCDD,

Table 2

Concentration of PCDDs, PCDFs, and PCBs and homolog totals in the spent foundry sands (n = 1).

Congener	TEF ^a	Spent foundry sand (ng kg ⁻¹)									
		1	2	3	4	5	6	7	8	9	10
2,3,7,8-TCDD	1	<0.02	0.03	<0.01	<0.02	<0.05	0.02	<0.03	0.02	0.14	0.07
1,2,3,7,8-PeCDD	1	0.03	0.13	0.02	0.03	<0.04	0.07	<0.03	0.15	0.72	0.24
1,2,3,4,7,8-HxCDD	0.1	0.02	0.09	<0.02	0.01	0.02	<0.02	<0.04	0.16	0.58	0.21
1,2,3,6,7,8-HxCDD	0.1	0.05	0.60	0.02	0.02	0.05	0.18	<0.04	0.21	0.81	0.33
1,2,3,7,8,9-HxCDD	0.1	0.03	0.35	<0.02	0.03	0.06	0.13	<0.04	0.15	0.66	0.23
1,2,3,4,6,7,8-HpCDD	0.01	0.38	5.3	0.42	0.15	0.60	0.74	0.21	1.2	5.0	1.6
1,2,3,4,6,7,8,9-OCDD	0.0003	27.8	44.8	2.9	1.6	8.8	5.9	3.0	3.0	12.5	2.4
2,3,7,8-TCDF	0.1	0.03	0.46	0.03	0.03	0.16	0.09	0.01	0.13	1.7	0.45
1,2,3,7,8-PeCDF	0.03	0.03	0.19	0.01	0.01	0.07	0.13	0.02	0.15	1.5	0.46
2,3,4,7,8-PeCDF	0.3	0.04	0.29	0.01	0.01	0.08	0.20	<0.04	0.21	2.6	0.72
1,2,3,4,7,8-HxCDF	0.1	0.06	0.25	0.01	<0.01	0.10	0.18	<0.04	0.18	2.3	0.63
1,2,3,6,7,8-HxCDF	0.1	0.04	0.18	0.01	0.01	0.04	0.15	<0.03	0.15	2.3	0.56
2,3,4,6,7,8-HxCDF	0.1	0.04	0.22	0.01	<0.01	0.02	0.24	<0.03	0.17	2.3	0.55
1,2,3,7,8,9-HxCDF	0.1	<0.02	<0.03	<0.02	<0.01	<0.03	<0.02	<0.03	<0.02	<0.04	<0.02
1,2,3,4,6,7,8-HpCDF	0.01	0.17	1.0	0.13	0.02	0.11	0.48	0.14	0.73	9.9	1.7
1,2,3,4,7,8,9-HpCDF	0.01	0.03	0.11	<0.02	<0.02	0.03	0.06	0.17	0.06	0.50	0.10
1,2,3,4,6,7,8,9-OCDF	0.0003	0.12	1.5	0.48	0.09	0.16	0.36	0.16	0.26	3.1	0.26
PCB-77	0.0001	0.30	47.4	0.43	2.0	7.1	2.1	0.53	0.81	4.4	1.2
PCB-126	0.1	0.12	1.2	0.02	0.06	0.24	0.72	0.01	0.22	2.0	0.38
PCB-169	0.03	0.02	0.09	<0.01	0.02	<0.03	0.06	<0.02	0.05	0.68	0.12
Sum TCDD		0.33	1.4	0.01	0.22	0.58	2.8	0.24	9.8	21.8	9.6
Sum PeCDD		0.33	1.4	0.00	0.17	0.42	1.5	0.83	8.4	20.7	9.7
Sum HxCDD		0.42	5.0	0.07	0.23	0.90	2.2	0.42	8.1	22.7	9.6
Sum HpCDD		0.76	10.3	0.63	0.35	1.5	1.5	0.44	2.7	10.2	3.5
Sum TCDF		0.66	5.1	0.33	0.50	1.6	5.3	0.08	6.1	53.0	16.8
Sum PeCDF		0.55	2.8	0.15	0.16	0.57	2.9	0.21	3.3	32.8	9.3
Sum HxCDF		0.46	2.2	0.14	0.10	0.45	1.5	0.37	1.9	22.1	5.6
Sum HpCDF		0.28	2.1	0.36	0.06	0.25	0.78	0.32	0.94	12.1	2.1

<, Below the method detection limit.

^a Values assigned by the World Health Organization (Van den Berg et al., 2006).

Table 3

A comparison of PCDD and PCDF concentrations in the spent foundry sands and background soils.

Congener	(This study)		Andersson and Ottesen (2008)		Schmid et al. (2005)	
	Min (ng kg ⁻¹)	Max (ng kg ⁻¹)	Min (ng kg ⁻¹)	Max (ng kg ⁻¹)	Min (ng kg ⁻¹)	Max (ng kg ⁻¹)
2,3,7,8-TCDD	<0.01	0.14	0.04	0.64	<0.05	0.7
1,2,3,7,8-PeCDD	<0.04	0.72	0.05	2.5	0.19	4.8
1,2,3,4,7,8-HxCDD	<0.02	0.58	<0.01	4.4	0.18	1.9
1,2,3,6,7,8-HxCDD	<0.04	0.81	<0.01	5.0	0.22	4.7
1,2,3,7,8,9-HxCDD	<0.02	0.66	<0.01	5.6	0.26	2.3
1,2,3,4,6,7,8-HpCDD	0.15	5.3	0.12	77.0	3.8	36.0
1,2,3,4,6,7,8,9-OCDD	1.6	44.8	1.2	830	22.0	120
2,3,7,8-TCDF	0.01	1.7	0.02	9.0	0.32	7.3
1,2,3,7,8-PeCDF	0.01	1.5	<0.01	6.4	0.23	6.6
2,3,4,7,8-PeCDF	<0.04	2.6	<0.01	10.0	0.34	8.1
1,2,3,4,7,8-HxCDF	<0.01	2.3	<0.01	9.6	0.47	6.6
1,2,3,6,7,8-HxCDF	<0.03	2.3	<0.01	6.0	0.29	6.2
2,3,4,6,7,8-HxCDF	<0.03	2.3	<0.01	4.9	<0.14	1.8
1,2,3,7,8,9-HxCDF	<0.01		<0.01	9.3	0.42	6.5
1,2,3,4,6,7,8-HpCDF	0.02	9.9	0.22	46.0	0.80	16.0
1,2,3,4,7,8,9-HpCDF	<0.02	0.50	0.06	8.6	<0.31	3.4
1,2,3,4,6,7,8,9-OCDF	0.09	3.1	0.31	66.0	3.1	51.0

and OCDD (Andersson and Ottesen, 2008). A comparison of the PCDD data acquired in this study, with the results obtained in the above mentioned studies, shows that SFSs contain PCDD concentrations at the lower end of the range found in background soils (Table 3).

The PCDF congener concentrations ranged from <0.01 to 9.9 ng kg⁻¹ in the SFSs (Table 2). Sand 9 (green sand from a steel foundry), contained the highest concentrations of each PCDF congener, which were an order of magnitude greater than in the other SFSs. While 1,2,3,4,6,7,8-HpCDF was detected at the highest concentration of 9.9 ng kg⁻¹, the TEF for this congener is 0.01, which is 10 times lower than the TEF for HxCDFs and TCDF. 2,3,7,8-TCDF was detected at concentrations from 0.03 to 1.7 ng kg⁻¹; the HxCDFs were found at concentrations ≤2.3 ng kg⁻¹. The PCDF congeners in the SFSs were all at concentrations below the maximum concentrations found in background soils (Table 3).

PCBs, which are synthetically derived contaminants, were also detected in the SFSs (Table 2). In this study, we only analyzed for PCB-77, -126, and -169, and did not take into account the other nine PCB congeners recognized by the WHO (Van den Berg et al., 2006). In contrast, however, the TEFs for PCB-126 and -169 are much higher at 0.1 and 0.03, respectively, compared to the TEF of 0.0001 for PCB-77 and 0.0003 for PCB-81 and the remaining mono-*ortho* PCBs (i.e. -105, -114, -118, -123, -156, -157, -167, and -189). The data show that the PCB concentrations in the SFSs decrease as the number of chlorine atoms increases on the congener. For example, the average concentration for PCB-77, -126, and -169 in the SFSs was 6.6, 0.5, and 0.15 ng kg⁻¹, respectively.

The contribution of various homologs of PCDDs and PCDFs to the total dioxin concentrations are presented at the bottom of Table 2. These homologs do contribute to the total dioxin load in the

samples, but have not been assigned a TEF as a result of their low toxicity compared to the congeners selected by the WHO. In many of the sands there was a fairly even distribution of the T, Pe, Hx, and Hp-CDD/F homologs. In a few sands, however, lower weight homologs were found at greater concentrations (e.g. sand 8, 9, and 10), except sand 2 (iron green sand) where the reverse trend occurred with the PCDD homologs. Sand 9 had the greatest PCDD/PCDF homolog total of 195 ng kg⁻¹, compared to 1.7 ng kg⁻¹ in sand 3. Similarities among the homolog concentration patterns in many of the SFSs may indicate a single source of the contamination, although the PCDD/PCDFs may be formed during the casting process. In the latter case, similarities may occur as a result of comparable casting conditions among foundries.

Table 4 shows the PCDDs, PCDFs, PCBs, and total concentrations corrected for their TEFs and expressed as TEQs. However, since we did not measure PCB-81 and mono-*ortho*-substituted PCBs, the PCB contribution to the total TEQ concentration is not known. Total concentrations ranged from 0.01 to 3.1 ng TEQ kg⁻¹, with an average concentration of 0.58 ng TEQ kg⁻¹. Foundry sand TEQ concentrations are similar to those obtained by Rogowski and Yake (2005), who analyzed forest, open, urban, and agricultural soils (*n* = 84) from Washington state. The average concentrations reported in that study were 2.3, 1.0, 4.1, and 0.14 ng TEQ kg⁻¹, respectively. These values were calculated using 1989 US EPA TEFs and, therefore, the TEQs are only about 4% higher than if determined using the 2005 WHO TEFs. The greatest total concentration of 3.1 ng TEQ kg⁻¹ was found in sand 9, a steel green sand. This concentration is about 100 times lower than the 300 ng TEQ kg⁻¹ limit considered by the US EPA for biosolids (US EPA, 2002). In sand 9, 23%, 25%, and 22% of the TEQ was attributed to 1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, and HxCDFs. Only 5% of the TEQ could be

Table 4

Toxicity equivalents (TEQs) of PCDDs, PCDFs, PCBs, and total dioxins in the spent foundry sands.

	Spent foundry sand (ng TEQ kg ⁻¹)									
	1	2	3	4	5	6	7	8	9	10
PCDDs	0.05	0.33	0.02	0.03	0.02	0.13	0.00	0.23	1.1	0.40
PCDFs	0.03	0.22	0.01	0.01	0.06	0.14	0.01	0.14	1.8	0.47
PCBs ^a	0.01	0.13	0.00	0.01	0.03	0.07	0.00	0.02	0.22	0.04
PCDD/PCDFs	0.08	0.55	0.03	0.04	0.08	0.27	0.01	0.37	2.9	0.87
Total ^{a,b}	0.10	0.68	0.04	0.05	0.11	0.34	0.01	0.40	3.1	0.91

^a Does not include PCB-81 and mono-*ortho*-substituted PCBs.

^b Sum of the PCDDs, PCDFs, and PCBs.

attributed 2,3,7,8-TCDD. Other spent sands with higher TEQs were sands 2 and 10 (iron and steel green sands), at 0.68 and 0.91 ng TEQ kg⁻¹, respectively. In sand 2, 49%, 32%, and 19% of the TEQ was attributed to PCDDs, PCDFs, and PCBs, respectively. In sand 10, 44%, 51%, and 5% was attributed to the TEQ, respectively. In the remaining SFSSs, PCDDs and PCDFs accounted from 76% to 94% of the total TEQ.

4. Conclusions

PCDD/PCDFs and PCBs can be detected in SFSSs, but were found at concentrations near the low end range in background soils. Because the molding sand components are mined from the environment, the source of the PCDD/PCDFs and PCBs could be associated with natural and/or anthropogenic pollution. However, the formation of these compounds (except PCBs) during the casting process cannot be excluded, but it is unlikely since SFSSs contain low levels of chlorine and organic carbon. Based on the low PCDD/PCDF and PCB concentrations in SFSSs, long-term soil applications should not result in excessive loadings (i.e. application of background soil concentrations to background soils cannot cause important accumulation in soil). It is also well documented that PCDD/PCDFs and PCBs are strongly bound in soils with little translocated into edible plant tissues (Hülster and Marschner, 1993; Simonich and Hites, 1995; Welsch-Pausch et al., 1995), although soil ingestion by grazing livestock is an important pathway if soil concentrations are substantially increased (Jones and Stewart, 1996; Wittsiepe et al., 2007). Overall, it is unlikely that spent sands from iron, aluminum, and steel foundries will present a risk to environmental receptors when beneficially used in soil-related applications.

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